Fine particles generated from the combustion of fossil fuels: Physicochemical characterization and direct inhalation toxicity studies at EPA

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Summary

In experiments at the U.S. Environmental Protection Agency's (EPA's) Environmental Research Center, heavy fuel oil was burned in a combustor simulating large industrial or utility boilers. Exhaust gases and particulate matter (PM) smaller than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) were diluted and sent to exposure chambers where groups of laboratory animals underwent short-term, direct inhalation exposure to $PM_{2.5}$ plus flue gases, or to filtered ambient air as a control. Measurements of particle size, concentration, and composition were compared to biological markers of pulmonary injury for each of the three groups to identify particle characteristics that may be responsible for adverse health effects.

Introduction

Numerous epidemiological studies have found an association between elevated levels of ambient $PM_{2.5}$ and measures of adverse health effects including premature mortality.[1-3] Toxicological studies of causal mechanisms have also found adverse biological reactions to $PM_{2.5}$ from specific sources, including residual fuel oil fly ash. To further investigate these causal mechanisms, a series of studies by the EPA's National Risk Management Research Laboratory (NRMRL) and the National Health and Environmental Effects Research Laboratory (NHEERL) were begun. These studies combined NRMRL's understanding of the physico-chemical characteristics of combustion-generated particles with NHEERL's expertise in the field of cardiopulmonary toxicity.

Experimental Approach and Setup

NRMRL's research combustors are used to simulate a range of combustion conditions found in full-scale combustion equipment. These tests focused on PM generated from residual oil combustion in simulated large utility and industrial waterwall furnaces and a small firetube boiler.[5]

Flue gases from these units were diluted with filtered ambient air to achieve the desired particle concentration (see Ref. 6). The dilution levels were between about 50:1 and 100:1, resulting in exposure PM concentrations of 2-5 mg/m^3, significantly higher than typical ambient concentrations but within the range of concentrations for these types of experiments. Particles larger than 2.5 μm in aerodynamic diameter were removed using a cyclone prior to entering into the dilution sampler. Post-dilution concentrations of nitrogen oxides (NO_x) and sulfur dioxide (SO_2) were initially estimated to be low enough (5-10 ppm for NO_x, 10-20 ppm for SO_2) to eliminate the need for additional gas treatment. Post-dilution carbon monoxide and oxygen concentations were well within the bounds required by animal testing protocols.

Diluted gases were ducted to a 1 m^3 whole-animal exposure chamber in an enclosure insulated to control temperature, noise, and humidity. A control chamber filled with filtered ambient air was also located inside the enclosure. Laboratory animals were transported directly from the animal facility and inserted into the chambers. After exposure, animals were transported back to the toxicology laboratories for sacrifice and testing for a broad range of cardiac and pulmonary damage markers. Healthy rats and mice were used to study animal-specific responses, and compromised animals (spontaneously hypertensive rats and mice exposed to *Streptococcus zooepidemicus*) were exposed to examine the response of animals with pre-existing health conditions.

Results

Seven series of tests have been conducted. Relatively modest adverse health impacts were noted with the first six test series. Further investigation concluded that excessive chamber SO_2 concentrations (40-50 ppm),[7] resulted in nasal passage inflammation and reduced airflow into the lungs, reducing the animals' pulmonary particle doses.

Approximately 90% of the SO_2 was then removed with a denuding system that used a sodium bicarbonate solution. Measurements of PM concentrations and size distributions at the denuder inlet and exit indicated an increase in particle concentration across the denuder, probably associated with formation of sulfate particles. Chemical analysis indicated no loss of combustion-generated particles across the denuder.

Prior to installation of the SO_2 denuder, mice exposed to diluted combustion gases for 4 hrs were tested for acute lung injury and susceptibility to *Streptococcus zooepidemicus* infection immediately and 24 hr after exposure. Exposure to the diluted flue gases caused a significant increase in neutrophils and lactate dehydrogenase in bronchoalveolar lavage but did not affect protein levels. Mortality to infection was low in all animals and was not affected by exposure to the combustion flue gases in these tests.[7]

Conclusions

The toxicological results to date are likely to have been influenced by excessive SO_2 concentrations. Further studies are being conducted with the denuder system to isolate the effects of particles alone. Although residual fuel oil represents a small fraction of fuel use, the mechanisms governing formation of metal-containing particles are the same as those for other fossil fuels containing trace metals. Thus, these studies provide a simple system for study of particle formation and related health effects. This work is a means to identify the health impacts of source-specific PM exposure, and can be extended to identify the impacts associated with specific particle characteristics. Future work may include direct exposure to PM generated from coal combustion in boilers, to particles from diesel and gasoline engines and from residential wood combustion, and to particles from combinations of these sources. Chronic exposure studies may also be conducted in future tests.

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